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Novel design of cyclic membrane gas separation process

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The analysis of membrane gas separation processes under transient conditions for practical applications is a rather unexplored domain. One of the main advantages of membrane processes compared to other separations is their ability to work under steady conditions, without any separate regeneration step. Nevertheless, unique separation performances can be obtained in some cases when a transient regime is applied. To our knowledge, Paul (1971) explored this issue in a pioneering study based on a cyclic operation. Recently existing cyclic processes have been reviewed by Wang et al. (2011b). According to the duration of the high pressure stage and the duration of the complete cycle, cyclic processes have been classified into short and long class.

Short class processes are based on Paul's initial idea. In general, one cycle is divided into two stages: high and low pressure stages. In a short class process, the high pressure stage duration must be comparable to the time-lag in order to use the large difference of the diffusion rates in transient states of different gas components. A permeate enriched in desired gas is thus obtained during this stage. After that, a low pressure stage when the membrane is being regenerated is not only compulsory but also very long with respect to the high pressure stage. By repeating the cycles, an extreme pure permeate can be achieved with respect to conventional steady-state operations based on the same membrane. Nevertheless, it has been shown by means of some theoretical and experimental studies that the productivity of such a process is too low and there is no separation effect in retentate. These unavoidable drawbacks imply that the short class processes might only be interesting for separation of specific gases.

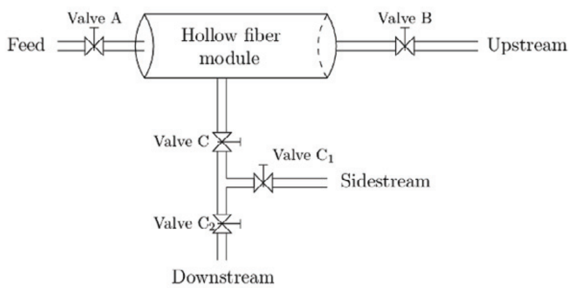

Figure 1 : Patented cyclic process design

Wang et al. (2011b) indicated the fundamental difference between short and long class processes: in short class processes, the transient behavior in the selective layer of the membrane is utilized, whereas in the long class processes, the transient behaviors occur in the system outside the membrane. Within the frame of a systematic cyclic process study, a new process (Fig. 1) is designed then patented (Wang et al. 2011a) in order to lower the drawbacks of Paul's design and achieve a competitive cyclic process. In this process, the transient

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behaviors in the system outside the membrane are essentially utilized and thus classified as a long class process.

This process is composed of a classical hollow fiber module with five automatic valves and some connections. Two options have been proposed according to operating complexity: basic option and sidestream option. Initially, the whole module is emptied and all valves are switched off. Once the process is launched, the module will be fed to a given pressure by opening valve A. Once the pressure set point is reached, valve A is closed then the diffusion occurs through the fibers. Then, two options are available:

- Basic option. Valves C_1 (always closed) and C_2 (always open) is not in use. Consequently, the sidestream flow does not exist. Valves B and C are opened at this stage in order to transport the gases in upstream and downstream to corresponding tanks. Due to the transport, the whole module is emptied. After that, all valves are closed and the cycle is finished.

- Sidestream option. All valves are in use. Valves C and C_1 are opened at this stage, a sidestream flow is collected in the corresponding tank. Then both valves C and C_1 are closed again in order to resume the diffusion through the fibers. After that, in Transfer stage, Valves B, C and C_2 are opened in order to transport the gases in upstream and downstream sides to corresponding tanks, due to the transport, the whole module is emptied. Then all valves are closed and the cycle is finished.

According to our simulation study, there is no more need to maintain a long regeneration stage for the novel process and the high pressure duration is significantly enlarged. Consequently, the patented process provides in general more than 1000 times of the production ability of a short class process under similar conditions. Then, based on gas compositions in different flux shown in Tab. 1, a systematic comparison between the novel design and conventional steady-state operations has been performed. Fig. 2 shows two types of comparison between the patented process with both options and conventional steady-state processes using stirred or cross flow modules.

	O_2 (slow gas) enriched flux	CO_2 (rapid gas) enriched flux
Conventional operations	Retentate	Permeate
Basic option	Upstream	Downstream
Sidestream option	Upstream	Sidestream

Table 1: Comparison of fluxes for conventional operations and both options

of the patented process

A great performance (purity of desired gas and its recovery ratio defined by recovered desired gas quantity divided by its quantity in feed) is achieved by increasing downstream/upstream volume ratio of the membrane module. For a volume ratio larger than 100, the patented process becomes in general more efficient than conventional steady-state operations. Furthermore, if the sidestream option is adopted, the process efficiency will be improved again and more degrees of freedom in operations will be available. For example, by a proper design, the downstream flow can be recycled with a composition close to the feed.

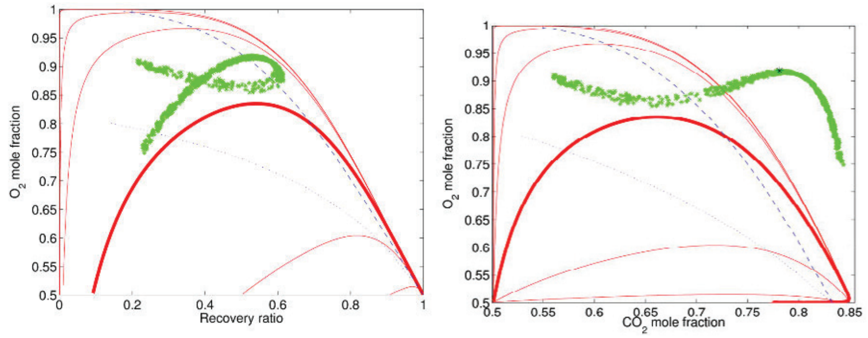

Figure 2 : Comparison of process performances. Left: O_2 recovery ratio of O_2 enriched flux in function of O_2 mole fraction in the same flux (Tab. 1). Right: CO_2 mole fraction in CO_2 enriched flux in function of O_2 mole fraction in O_2 enriched flux (Tab. 1). Continued red lines: patented process using basic option with different volume ratios γ (0.1, 1, 10, 100, 1 000 and 10 000 from bottom to top respectively). Bold continued red line: γ = 10. Dashed blue line: conventional operations using cross flow modules. Dotted blue line: conventional operations using stirred modules. Green stars: patented process using sidestream option with γ = 10 and the downstream flux is recycled. $\alpha(CO_2/O_2)$ = 5.8, O_2 mole fraction in feed = 0.5

For the black star marked in Fig. 2 (right), the recycle ratio defined by downstream flux divided by feed flux is 20.4 %. By applying corresponding operating conditions, one flux with 90% O_2 and another with 78% CO_2 can be obtained at the same time, which is impossible in the case of conventional steady-state operations.

The present communication intends to provide a complete analysis of operation possibilities of such a process and make a systematic comparison to conventional operations. Furthermore, an experimental proof has been performed in the LRGP (Nancy) which gives a strong support to the process design.

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